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Baseline of butyltin contamination in sediments of Sundarban mangrove wetland and adjacent coastal regions, India

Blanca Antizar-Ladislao,[*,a] *Santosh Kumar Sarkar,*[*,b] *Peter Anderson,*[c] *Tanya Peshkur,*[c] *Bhaskar Deb Bhattacharya,*[b] *Mousumi Chatterjee,*[b] *and Kamala Kanta Satpathy.*[d]

[a] Institute for Infrastructure and Environment, School of Engineering, University of Edinburgh, William Rankine Building, Edinburgh, United Kingdom EH9 3JL, United Kingdom. Fax: +44131 6506781; Tel: +44131 6505712; E-mail: B.Antizar-Ladislao@ed.ac.uk

[b] Department of Marine Science, University of Calcutta, 35 Ballygunge Circular Road, Calcutta 700019, India. E-mail: cusarkar@gmail.com

[c] Scottish Environmental Technology Network (SETN) Royal College Building, Faculty of Engineering, University of Strathclyde, Glasgow, G1 1XW, Scotland, United Kingdom.

[d] Indira Gandhi Centre for Atomic Research, Environmental and Industrial Safety Section, Safety Group, Kalpakkam 603102, Tamil Nadu, India.

Abstract

The study reports the first assessment for the quantification and speciation of butyltins in surface marine sediment samples (0-5 cm) from intertidal mudflats of Sundarban mangrove wetland along with the Hugli (Ganges) river basin, eastern coastal part of India. Concentrations of tributyltin (TBT), dibutyltin (DBT) and monobutyltin (MBT) were monitored at 16 stations and present at all study areas, in concentrations in sediments up to 84.2 ng g^{-1}, 26.4 ng g^{-1} and 48.0 ng g^{-1} of TBT, DBT and MBT, respectively. Significant correlations were obtained between MBT and DBT ($r=0.62$, $p=0.01$) and DBT and TBT ($r=0.54$, $p=0.03$). Calculated Butyltin Degradation Index (BDI) values indicated recent contamination of butyltins at 8 stations, and suggested either no degradation of TBT or very recent degradation at a 4 further stations. Additionally, BDI values also indicated no recent inputs of butyltins in 4 stations (only MBT present in one of these stations). High concentrations of butyltins, particularly TBT, have the potential to induce ecotoxicological impacts based on levels specified in Australian Sediment Quality Guidelines (SQGs). This study indicated that the majority of the analyzed stations were in the highest range of priority, due to high TBT concentrations.

Keywords: Butyltins, Sediment, Sundarban, India.

INTRODUCTION

Since the 1960s, organotin-based antifouling paints have been widely applied on the hulls of ships and boats and on surfaces that are in prolonged contact with seawater (Sonak *et al*. 2009). This controls the growth of fouling organisms such as barnacles, mussels, oysters, and tubeworms, but also being described as the most toxic substance ever introduced into the marine environment (Goldberg 1986), have produced a detrimental environmental impact (Antizar-Ladislao 2008). In fact, it is currently being investigated whether organotins might behave as endocrine disruptors (Sumpter 2005; Lagadic *et al*. 2007; Nakanishi 2008; Oehlmann *et al*. 2007).

This has led to national and international regulations of TBT. In 2001, the Anti-Fouling Systems (AFS) Convention that prohibits the use of harmful organotins in antifouling paints used on ships was adopted, and subsequently, the International Maritime Organization (IMO) called for a global treaty that bans the application of TBT-based paints starting 1 January 2003, and total prohibition by 1 January 2008, in awareness of its undesirable effects (Sonak *et al*. 2009; IMO 2001; CD 2002). India is neither a signatory to the AFS Convention 2001 nor does it have legislation prohibiting the use regarding TBT-based paints on ship hulls. However, it is a signatory to many international conventions for the protection of the marine environment. At the national level, India has The Hazardous Wastes (Management and Handling) Rules, 1989 (as amended in 2003), with guidelines for the manufacture, storage, and import of hazardous waste as well as for import and export of hazardous waste in the country. Amendment to this Act has widened the definition of 'hazardous waste', which now includes a number of compounds or substances including organotin compounds (Sonak *et al*. 2009).

Major ship-breaking yards are in Asian countries such as India, Pakistan, China and Bangladesh. Thus, contamination due to the presence of organotins in sediments and waters in India is expected. While a relative large number of studies have involved surveys on organotin distribution in sediments in many countries, to date, a limited

number of studies have focused on the occurrence of organotins in sediments in India (Jadhav *et al*. 2009; Meena *et al*. 2009; Mukherjee *et al*. 2009).

Thus the objective of this study was to determine the concentration of TBT and its degradation products, DBT and MBT, in coastal sediments covering 16 key regions of Sundarban mangrove wetland and along the stretch of the Hugli River affected by different intensities of maritime traffic activities within the context of reported values from other countries and with reference to available sediment quality and ecotoxicological standards.

EXPERIMENTAL

Site description

Hugli river - The Hugli river (~260 km long), a major distributory of the Ganges river in the east coast of India. It provides perpetual supply of water to the plains of West Bengal for multipurpose activities, such as, irrigation, navigation of small ships and fishing boats, human and industry consumption together with fishing. Along the stretch of the river, there are small jetties that are used for berthing of small fishing vessels and barges. Both the banks of the river in its downstream stretch are used for different large and small scale electrical, jute, brick, paper and pulp, electrochemical and petrochemical industries. The bank of the river is also used as several burning ghats, the wastes of which are frequently discharged directly to the river system.

Sundarban - The deltaic region formed at the estuarine phase of Hugli river is famous for its luxuriant mangrove vegetation, known as Sundarban, and acclaimed as UNESCO World Heritage Site for its capacity of sustaining an excellent biodiversity. The area is interspersed with a large number of islands and tidal channel systems through which semidiurnal tides of meso- macro-tidal amplitude interplay with moderate to strong wave effects. The wave and tide climate of this low-lying tropical coast primarily controls the sediment dispersal patterns. Biogenic sub-duction and re-

suspension also play a significant role in the vertical and lateral transportation of sediments.

Sampling

Surface marine sediments were collected during winter season (January–February 2009) using a grab sampler during low tide from 16 stations along the stretch of Hugli river as well as Sundarban mangrove wetland, West Bengal, eastern coastal part of India. Sampling locations were selected considering the sediment dispersal patterns along the drainage network systems (Fig.1, Table 1) and suspected presence of butyltins as a consequence of boating activities. The stations are representative of the variable environmental and energy regimes that cover a wide range of substrate behaviour, wave-tide climate, intensity of bioturbation (animal-sediment interaction), geomorphic-hydrodynamic regimes and distances from the sea, Bay of Bengal.

Sediment samples were collected in triplicate from the top 3-5 cm of the surface at each sampling site, pooled and thoroughly mixed. Immediately after collection, the samples were placed in sterilized plastic bags in the ice box and transported to the laboratory. Samples were oven dried at 50°C, gently disaggregated, and individually transferred into pre-cleaned inert polypropylene bags and stored in deep freeze prior to analysis. The 16 pooled samples were each divided into two aliquots: one unsieved, for the determination of sediment quality parameters and another one sieved through 63 µm metallic sieves, for butyltin analysis.

Physical and chemical analysis of sediments

The sediment samples were characterized for particle size, pH, and organic carbon. Particle size was determined by sieving the dried samples in a Ro-Tap Shaker (Krumbein and Pettijohn 1938) manufactured by W.S. Tyler Company, Cleveland, Ohio and statistical computation of textural parameters was done by using the formulae of Folk and Ward (1957). The determination of pH was conducted with a digital pH

meter 101E Deluxe Model (EI Products, India) using a combination glass electrode manufactured by M.S. Electronics (India) Pvt. Ltd. Organic carbon (C_{org}) content of the soil was determined following a rapid titration method (Walkley and Black 1934) after oxidizing with potassium dichromate ($K_2Cr_2O_7$) in presence of sulphuric acid (H_2SO_4). Samples (5.0 g) of sieved sediment were weighed into 25-ml beakers and spiked with 150 µl of the surrogate, triphenyl tin chloride (TPT), resulting in a concentration of 3 µg g^{-1}. The spiked sediments were then thoroughly mixed with 15-20 g of analytical sand (previously heated for 4 hours at 550^{o}C), transferred to 34 ml accelerated solvent extraction (ASE) stainless-steel extraction cells and set aside for two hours prior to extraction. Samples were extracted using an ASE 300 solvent extractor (DIONEX, Camberley, UK) using a method (100^{0}C x 25min) based upon DIONEX Application note 339 (DIONEX 2001).

Extracts were transferred to 250 ml volumetric flasks containing 7.3 g of sodium chloride (NaCl), diluted with high performance liquid chromatography (HPLC) grade water, and then pH-adjusted to 5.0 ± 0.1 with 1 M sodium hydroxide (NaOH). One millilitre of a 5 % w/v sodium tetraethylborate ($(C_2H_5)_4BNa$) aqueous solution (derivatizing agent) was added to each flask and the volume was adjusted to 250 ml with HPLC grade water. Samples were then transferred into 500 ml glass jars. Finally, 3 ml of *n*-hexane was added and the samples were shaken for 12 hrs on a Laboshake end-over end shaker (Gerhardt, Brackley, UK). After 12 hrs, shaking samples were transferred back into 250 ml volumetric flasks, left for 15-30 min to settle and a 1 ml aliquot of the *n*-hexane layer was transferred into a 2 ml GC vial and spiked with 10 µl of internal standard, tetrabutyltin (TeBT), resulting in a concentration of 10 mg l^{-1}. The stock standard solution had been prepared in methanol and contained 10,000 mg l^{-1} of each individual butyltin (BT) compound [monobutyltin trichloride, MBT (201057-5G, Sigma-Aldrich); tributyltin chloride, TBT (T50202-5G, Sigma-Aldrich) and dibutyltin DBT (205494-50G, Sigma-Aldrich)]. Analytical standards of 0.1, 0.5 and 2 mg l^{-1} of the individual BT compounds were prepared by adding 30, 150 and 600 µl of intermediate stock standard (concentration of 10 mg l^{-1} BT each) to analytical sand and extracting under the same conditions as the sediments. Standard samples were spiked

with 150 µl of TPT (245712-5G, Sigma-Aldrich). Also, 10µl of TeBT (T6008-5G-A, Sigma-Aldrich) was added to 1 ml of *n*-hexane extracts.

Analysis of Sediment Extracts

Sediment extracts were analyzed on a Thermo Finnigan gas chromatograph (GC) equipped with a mass selective detector (MS), coupled to a Thermo Tri-plus auto-sampler (all Thermo Fisher, Loughborough, UK). A volume of 1 µl of each extract was introduced into an injection port held at a temperature of 250^{o}C and operated in the splitless mode with a splitless time of 2 minutes and split flow of 50 ml min^{-1}. The initial oven temperature was 60^{o}C for 4 minutes after which the oven was heated to 250^{o}C at 10^{o}C per minute and held for 4 minutes followed by another ramp of 10^{o}C per minute to 300^{o}C and held for 2 minutes. Separation was achieved using a 30m RTX-5 column with an internal diameter of 0.25 mm and film thickness of 0.25 µm (Restek, UK) maintained at a constant flow of 1.0 ml min^{-1} of helium. GC-MS interface temperature was set to 250°C, and detection was performed in the electron impact ionization mode and selected-ion monitoring (EI-SIM). The emission current was set to 150 µA and the acquisition rate to 2 scans per second with a detector voltage of 260 V. Correct identification and quantification of a given analyte was assured by using two compound specific ions and a mass ratio similar to the one determined with calibration (variation <10%).

QC/QA

All organotin standards, including surrogate and internal, were obtained from Sigma Aldrich (Gillingham, UK). All other reagents were obtained from Thermo Fisher UK (Loughborough, UK) and were of analytical grade. Analyses of organotins were conducted according to the standard operating procedures of the National Institute for Coastal and Marine Management/RIKZ (Stronkhorst *et al*, 2004) by GC-MS (split/splitless injection) and the limit of detection of each component was 1 ng g^{-1}. For all the analyses, blanks (solvent) and spiked blanks (standards spiked into solvent)

were routinely analyzed. The calibration was frequently checked during the analysis of samples by the repeated analysis of quality control standards. Recoveries of surrogate standards and internal standards in samples were above 80% throughout all sample analyses. Results of the standard reference materials used to ensure the QC/QA of the analysis are shown in Table 2.

Statistical analysis

Pearson product correlation coefficients between butyltins and organic carbon in sediment were calculated using STATISTICA (StatSoft 2011).

RESULTS AND DISCUSSION

Sediment geochemistry

The sediment quality parameters (pH, textural properties and C_{org}), as depicted in Table 3, were characteristically different from each other, and these variations were mostly reflected in the butyltin concentrations in sediments from the 16 investigated locations (Table 4). Values of pH ranged from slightly acidic to basic (6.5 to 8.8) where the acidic nature was exclusively recorded at Jharkhali S_{12}, most probably partly due to the oxidation of iron (II) disulphide (FeS_2) and iron (II) sulphide (FeS) to sulphate ion (SO_4^{2-}) and partly to the decomposition of mangrove litter and hydrolysis of tannin in mangrove plants which releases various kinds of organic acids (Liao 1990). Low organic carbon (C_{org}) values (< 1%) were recorded in all the stations excepting at Gushighata S_{15}, where the maximum value (1.04%) was found. The prevalent low C_{org} values were probably the result of sedimentation and mixing processes at the sediment water interface where the rate of delivery as well as the rates of degradation by microbial-mediated processes can be high (Canuel and Martens 1993). Very low C_{org} values in intertidal zone sediments in Sundarban have been also recorded in previous studies and related to the poor absorbability of organics in negatively charged quartz grains, which predominate in sediments in this estuarine environment (Sarkar *et al*. 2004; Chatterjee *et al*. 2007). Regarding textural composition, the 16 stations also exhibited wide variations, silty clay to sandy which are related to typical vigorous estuarine processes such as mixing, suspension, resuspension and flocculation.

Distribution of butyltins

Concentrations of MBT, DBT and TBT in marine surface sediments in West Bengal, India were monitored at 16 stations along the Hugli River and Sundarban (Table 4). Butyltins were found at all study areas, ranging from below detection limit to highest concentrations of TBT at S_{12} (84.2 ng g^{-1}), DBT at S_2 (26.4 ng g^{-1}) and MBT at S_{11} (48.0 ng g^{-1}). Butyltin concentrations within the same range were reported in North west Sicilian coast of Italy (7.2-66.0 ng g^{-1}) (Chiavarini *et al*. 2003), but considerably higher butyltin concentrations have been reported in coastal areas of Thailand and India with TBT concentrations in sediments up to 1,246 ng g^{-1} and 2,800 ng g^{-1} (Harino *et al*. 2006; Garg and Bhosle 2005), respectively, and in the sediments of the Zuari estuary, west coast of India with TBT concentrations in sediments up to 670 ng g^{-1} (Meena *et al*. 2009).

Out of the three butyltin species, TBT generally prevailed over DBT and MBT except at stations, S_1, S_{11}, S_{15}, (Table 4), where MBT was predominant particularly at Bakkhali S_{15}, where MBT contributed to more than 50% of total butyltin. This indicated that the concentration of TBT degradation product was considerable at this specific site (DBT (Sn): 23.3 ng g^{-1} d.w., MBT (Sn): 48.0 ng g^{-1} d.w.), while the parent compound was present at a lower concentration (TBT (Sn): 12.3 ng g^{-1} d.w.). DBT + MBT concentrations in this sample were about 6 fold higher than TBT concentrations, suggesting that TBT would have been degraded.

The results from this study make apparent that butyltin contamination is widely distributed along marine sediments in the investigated region in India. This is affected mainly by intensive use of antifouling paints to control growth of antifouling organisms on mechanized fishing boats and commercial ships along with the effluents from municipal sewage treatment plants. However, the prevalent contamination reveals lower concentrations than other coastal regions in India (e.g., Kochi Harbour, 16-16,816 ng g^{-1}, as Sn, d.w. and Mumbai Harbour, 4.5-1,193 ng g^{-1}, as Sn, d.w., south and west coast of India, respectively. It should be noticed that concentrations in ng g^{-1}, as Sn, are lower than those given as MBT, DBT, TBT by a factor of ca. 0.4).

Using the pollution classification proposed by Waite *et al*. (1991) the investigated stations in the lower stretch of Hugli river, including some stations in Sundarban may be ranked as follows: station S_{12} (Jharkhali) as medium contaminated (60-200 ng g^{-1} TBT) with the highest concentration of TBT (84.2 ng g^{-1}), as aforementioned, stations S_2-S_4, S_6-S_{11} and S_{13}-S_{16} as lightly contaminated (10-50 ng g^{-1} TBT) and stations S_1-S_5 (Diamond Harbour) as non-contaminated (0-10 ng g^{-1} TBT). According to Waite *et al*. (1991) sediment concentrations greater than 1,000 ng g^{-1} TBT are likely to contain paint particles.

The presence of DBT and MBT was indicative of TBT biotic and/or microbial degradation. According to previous studies, TBT degradation can be evaluated following a degradation index (BDI), which also assists in predicting whether butyltin contamination is recent or not (Diez *et al*. 2002). BDI was calculated as, BDI = [([DBT] + [MBT])/[TBT]], where [MBT], [DBT] and [TBT] refer to their concentrations. Values of BDI lower than 1 indicate that butyltin contamination is recent, and values of BDI higher than 1 indicate that there were no recent inputs of butyltins to the sediments. Thus, the calculated BDI in marine sediments in India analyzed in this study ranged from 0.57 to 2.50, which are in the same range as those reported in the Mediterranean Region in Spain within the range 0.15 to 2.94 (Diez *et al*. 2002), although sediments collected from station S_{11} presented a higher BDI value of 5.80.

Sediments collected from stations S_3, S_4 and S_{12} that presented a BDI lower than 1 are indicative of recent inputs of butyltins to the sediments in these stations. Additionally, stations S_9, S_{10}, S_{13} and S_{16}, presented concentrations of TBT in the range 17.9-32.4 ng g^{-1}, while DBT and MBT were not detected in those stations, suggesting that in these stations recent inputs of butyltins would have occurred or there was a low degradation rate. On the contrary, higher BDI values encountered in S_{11} were indicative of a high degradation rate.

The degradation products of butyltins are far less toxic than the parent compound (Fent 1996). Degradation is therefore, a process by which remediation of contaminated sediments can occur involving natural attenuation processes (Burton *et al*. 2006;

Antizar-Ladislao 2009). The observed values of butyltins indicate that these sediments are contaminated with butyltins and at some locations the contamination appears to be high enough to produce harmful effects on marine organisms (Waite *et al.* 1991).

Ecotoxicological concerns

In this study, the Australian Sediment Quality Guidelines (SQG) were used to assess the potential ecotoxicological impacts for TBT measured in marine sediments in West Bengal region, India. The Australian SQG reported for TBT are 12.5 and 175 ng TBT g^{-1} for low and high trigger values (ANZECC/ARMCANZ 2000). Out of the 16 sediment samples from the Hugli river and Sundarban mangrove wetland, TBT was not detected in one sample (S_5) and the rest contained TBT concentration higher than the high trigger value. Therefore the sediments from the majority of the sampling points are highly contaminated and may pose a threat to benthic biota like high larval mortality, several malformations of shells and reduced reproduction in oysters (Alzieu et al. 1991), growth retardation in mussels and microalgae (Salazar and Salazar 1991, Beaumont and Newman 1986), to mention a few.

Comparison of the OC-normalised TBT contents in sediments of Sundarban mangrove wetland (Table 4) against chronic toxicity LC_{10} (i.e., lethal concentration affecting 10% of the population for day 42) values for juvenile polychaete growth in a sediment with 0.6% OC showed that TBT contents exceeded the LC_{10} value of 5,879 ng TBT g^{-1} OC given by Meador and Rice (2001) in five stations (S_2-S_4, S_{11}-S_{12}). High concentrations of TBT in these five stations may thus affect local polychaete species. Based on the tissue residue approach, protection against severe adverse sublethal effects for many salmonoid prey species should be achieved with a TBT sediment concentration of 6,000 ng g^{-1} OC. The tissue residue approach is based on tissue concentrations as an alternative method to those using water concentrations for generating sediment quality guidelines (Meador *et al.* 2002). TBT concentrations in four stations (S_2-S_3, S_{11}-S_{12}) exceeded this value (6,000 ng g^{-1} OC), and therefore effects in salmonoid prey species occurs. It should be mentioned that responses to TBT may occur at lower concentrations that those reported earlier (Meador and Rice 2001; Meador *et al.* 2002) because of additive or synergistic effects produced by other contaminants bioaccumulated at the site.

Sorption of organotins to sediments is a fast and reversible process. While undisturbed sediments containing organotins in the lower layers should not be a major input source for such compounds in the overlaying water, any resuspension of heavily organotin contaminated sediments by turbulence (for example dredging activities, tides or storms), will lead to enhanced organotin concentrations in the water column (Berg *et al.* 2001). High correlations between silt fractions (particularly fine and very fine, although the grain-size *per se* did not appear to be the principal determinant of sorption) present in sediments and partitioning of TBT have been reported (Langston and Pope 1995). Particularly for sediments with a C_{org} concentration lower than 0.5% sorption to mineral phases such as silt becomes important. Furthermore, hydrophobic partitioning of organotins to non polar organic matter at the pH values recorded in this study will not be important (Burton *et al.* 2004). As fine and very fine silt is also most readily suspended by turbulence in the overlaying water, silty sediments will be involved in physical transport processes and will consequently facilitate organotin-transport processes. Due to the lipophilic and ionic properties of TBT, TBT levels will be influenced by the concentrations of organic carbon (Antizar-Ladislao 2008), and therefore correlations with organic carbon content are expected (Garg et al 2009). However correlations between the organic carbon for the marine sediments sampled in this study ranged from 0.04 to 1.04 (Table 3) and butyltins were found. It is thus suggested that particular attention should be given to sites with high concentrations of TBT (S_2-S_4, S_{11}-S_{12}), C_{org} values lower than 0.5% (S_3, S_5, S_8, S_{10}-S_{11}), high silt content (S_1-S_6, S_8-S_{10}, S_{12}-S_{16}): Bajbaj (S_3).

CONCLUSIONS

This is, to the knowledge of the authors, one of the first studies reporting butyltin concentrations in marine sediments in the Sundarban mangrove wetland including Hugli (Ganges) River basin, India. Butyltins were found at all study areas, within the same range reported in north west Sicilian coast of Italy, from below detection limit to highest concentrations of TBT at S_{12} (84.2 ng g^{-1}), DBT at S2 (26.4 ng g^{-1}) and MBT

at S_{11} (48.0 ng g^{-1}). Fishing and aquacultural activities at S_2-S_3, S_{11}-S_{12} are a concern, as the concentrations found in this station may pose harmful effects to molluscs and other marine organisms, and ultimately these persistent organic contaminants may enter the trophic chain. Station S3 is of particular concern as it also present low C_{org} values and high silt content, were butyltin compounds will be prone to mobilisation. Higher butyltin concentrations have been reported by previous studies in coastal areas of India (up to 2,800 ng g^{-1}). Thus the present observation encourages basic ecotoxicological research on native species of Indian subcontinent.

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Table 1 Sampling stations and description (Collection of marine sediment samples during January-February 2009).

Collecting Station		***Description of ecological stresses***
No[a] Location of the stations (S1- S_5*) (along the stretch of Hugli river)*		
1	Barrackpur (S_1)	Upstream, main stresses are industrial effluents, domestic sewage disposal, boating, bathing, occasional immersion of idols
2	Dakshineswar (S_2)	Upstream, main stresses are industrial and domestic effluents, recreational and traditional practices, bathing, boating, etc
3	Bajbaj (S_3)	Upstream, Industrial stress mainly power plant discharges, domestic sewage, boating, etc
4	Babughat (S_4)	Upstream, domestic and industrial effluents, bathing, frequent immersion of idols, boating etc
5	Diamond Harbour (S_5)	Downstream, mainly boating, recreational activities, bating etc
Location of the stations (S_6 – S_{16}) *in Sundarban coastal regions*		
6	Lot 8 (S_6)	Frequent dredging, boating, fishing etc
7	Kakdwip (S_7)	Boating, aquacultural and agricultural runoffs, fishing, dredging etc
8	Gangasagar (S_8)	Boating, tourist activities, dredging, fishing, agricultural, domestic and aquacultural practices
9	Chemagari (S_9)	Boating and fishing
10	Haribhanga (S_{10})	Boating and fishing
11	Bakkhali (S_{11}),	Tourist activities, boating and anthropogenic wastes
12	Jharkhali (S_{12})	Boating, fishing and aquacultural activities
13	Gosaba (S_{13})	Boating, fishing and anthropogenic activities
14	Canning (S_{14})	Boating and fishing
15	Gushighata (S_{15})	Effluents from east Kolkata tanneries, agricultural and other anthropogenic activities, boating
16	Dhamakhali (S_{16})	Effluents from tanneries, boating and fishing

[a] See Fig. 1 for sampling site location

Table 2 Results of the analyses of certified reference materials.

	MBT		**DBT**		**TBT**		**TeBT**	**TPT**		
	Conc. (mg kg^{-1})	**Recovery (%)**	**Conc. (mg kg^{-1})**	**Recovery (%)**	**Conc. (mg kg^{-1})**	**Recovery (%)**	**Recovery (%)**	**Recovery (%)**	**Weight of soil (g)**	**Volume of solvent (ml)**
057 Reference Material 1	0.14	**22.9**	0.47	**60.8**	0.45	**93.8**	**91.0**	-	1.10	3.0
057 Reference Material 2	0.11	**17.8**	0.37	**48.1**	0.38	**79.6**	**96.4**	-	1.01	3.0
057 Reference Material 3	0.08	**12.3**	0.39	**50.2**	0.44	**91.1**	**90.5**	-	1.10	3.0
057 Reference Material 4	0.16	**26.2**	0.45	**58.7**	0.47	**98.9**	**97.2**	-	1.02	3.0
057 Reference Material 5	0.17	**27.7**	0.46	**60.1**	0.43	**88.9**	**103.3**	-	1.02	3.0

	MBT	**DBT**	**TBT**
Average for 1g (n=5) sample size	**0.13**	**0.43**	**0.43**
SD for 1g sample size (n=5)	0.04	0.05	0.03
RSD (n=5)	29.63	10.74	7.88
Average Recovery (%) (n=5)	**21.4**	**55.6**	**90.5**
SD for 1g sample size (n=5)	6.34	5.97	7.13
RSD (n=5)	29.63	10.74	7.88

Table 3 Physicochemical properties of the sixteen sediment-sampling stations of marine sediments of the Hugli river and Sundarban mangrove wetland (India).

Collecting Station		**C_{org} %**	**pH**	**Sand (%)**	**Silt (%)**	**Clay (%)**
No	***Location***					
1	Barrackpur (S_1)	0.59	7.9	10.59	69.91	19.50
2	Dakshineswar (S_2)	0.52	7.8	1.21	78.40	20.39
3	Bajbaj (S_3)	0.40	7.6	11.98	85.20	2.82
4	Babughat (S_4)	0.57	7.9	0.97	86.74	12.29
5	Diamond Harbour (S_5)	0.41	8.4	11.61	84.12	4.27
6	Lot 8 (S_6),	0.78	8.3	1.15	84.82	14.03
7	Kakdwip (S_7)	0.66	8.8	6.18	5.74	88.08
8	Gangasagar (S_8)	0.41	8.1	25.97	63.53	10.48
9	Chemagari (S_9)	0.55	8.1	22.80	67.70	9.50
10	Haribhanga (S_{10})	0.40	8.1	23.65	62.05	14.30
11	Bakkhali (S_{11})	0.04	8.1	98.18	0.84	0.98
12	Jharkhali (S_{12})	0.81	6.5	13.04	68.36	18.60
13	Gosaba (S_{13})	0.67	8.7	0.68	48.66	50.66
14	Canning (S_{14})	0.83	8.0	2.10	67.63	30.27
15	Gushighata (S_{15})	1.04	8.0	11.85	30.85	57.30
16	Dhamakhali (S_{16})	0.72	8.1	1.00	85.80	13.20

[a] See Fig. 1 for sampling site location

Table 4 Concentration of monobutyltin (MBT), dibutyltin (DBT) and tributyltin (TBT) content in sediments (ng g^{-1} d.w.) from sixteen samples stations of coastal sediments of the Hugli river and Sunderban mangrove wetland (India).

					OC - Normalized concentration		
Sampling stations	***[MBT]***	***[DBT]***	***[TBT]***	***BDI= ([DBT]+[MBT])/[TBT]***	***[MBT]***	***[DBT]***	***[TBT]***
Barrackpur (S_1)	14.8	9.2	9.6	2.50	2,508	1,559	1,627
Dakshineswar (S_2)	20.3	**26.4**	44.7	1.04	3,904	5,077	8,596
Bajbaj (S_3)	16.0	7.7	25.9	0.92	4,000	1,925	6,475
Babughat (S_4)	18.1	14.1	32.9	0.98	3,175	2,474	5,772
Diamond Harbour (S_5)	14.8	<d.l.	<d.l.	<d.l.	3,610	<d.l.	<d.l.
Lot 8 (S_6)	20.4	16.1	22.6	1.61	2,615	2,064	2,897
Kakdwip (S_7)	15.9	13.5	18.4	1.60	2,409	2,045	2,788
Gangasagar (S_8)	26.0	<d.l.	14.3	1.82	6,341	<d.l.	3,488
Chemagari (S_9)	<d.l.	<d.l.	20.4	<d.l.	<d.l.	<d.l.	3,709
Haribhanga (S_{10})	<d.l.	<d.l.	17.9	<d.l.	<d.l.	<d.l.	4,475
Bakkhali (S_{11})	**48.0**	23.3	12.3	5.80	120,000	58,250	30,750
Jharkhali (S_{12})	22.6	25.2	**84.2**	0.57	2,790	3,111	10,395
Gosaba (S_{13})	<d.l.	<d.l.	28.9	<d.l.	<d.l.	<d.l.	4,313
Canning (S_{14})	22.6	7.1	22.6	1.31	2,723	855	2,723
Gushighata (S_{15})	38.5	10.7	32.8	1.50	3,702	1,029	3,154
Dhamakhali (S_{16})	<d.l.	<d.l.	32.4	<d.l.	<d.l.	<d.l.	4,500

d.l. detection limit (< 1.0 ng g^{-1} d.w.)

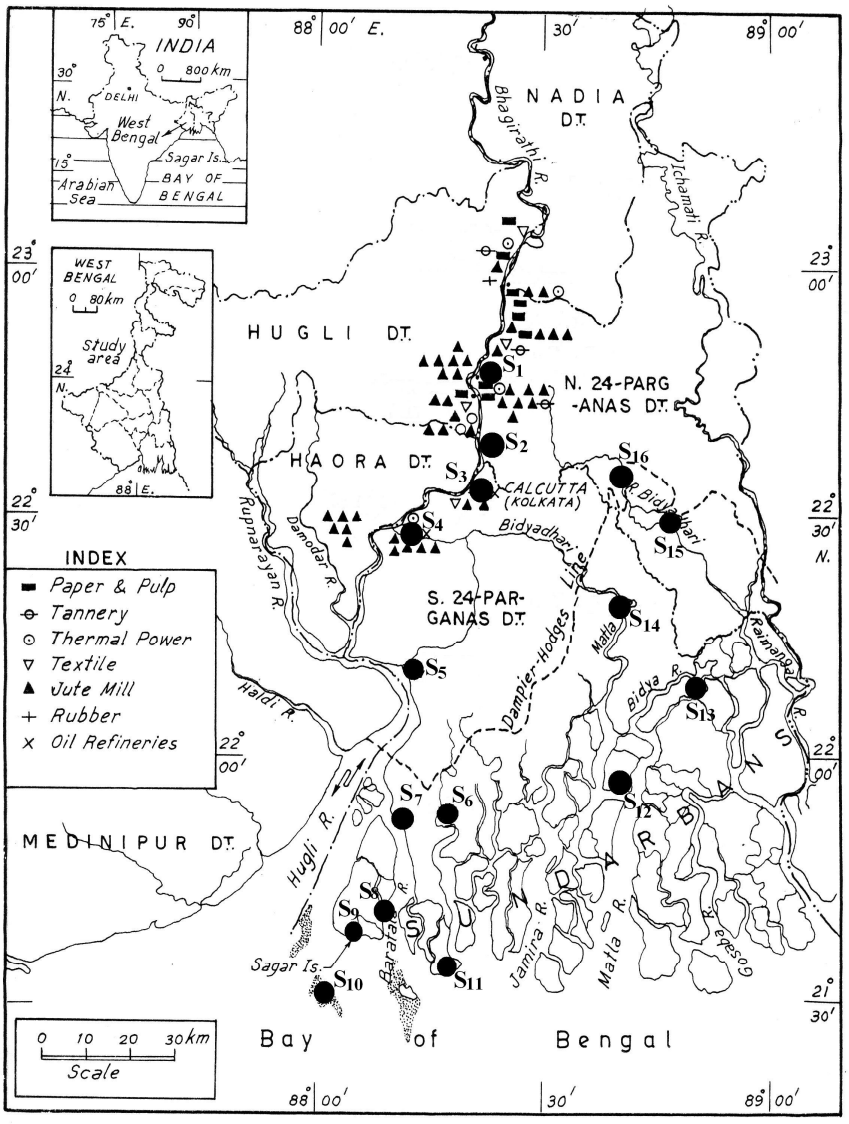

Fig. 1 Map showing location of the 16 sampling stations covering the Hugli river basin (S_1 - S_5) and the adjacent Sundarban mangrove wetland (S_6 - S_{16}) (See Table 1 for station description).